

Tar production from biomass pyrolysis in a fluidized bed reactor: a novel turbulent multiphase flow formulation

J. Bellan and D. Lathouwers ^a

^aJet Propulsion Laboratory, California Institute of Technology, 4800 Oak Grove Drive, MS 125/109, Pasadena, CA 91109, USA

A novel multiphase flow model is presented for describing the pyrolysis of biomass in a 'bubbling' fluidized bed reactor. The mixture of biomass and sand in a gaseous flow is conceptualized as a particulate phase composed of two classes interacting with the carrier gaseous flow. The solid biomass is composed of three initial species: cellulose, hemicellulose and lignin. From each of these initial species, two new solid species originate during pyrolysis: an 'active' species and a char, thus totaling seven solid-biomass species. The gas phase is composed of the original carrier gas (steam), tar and gas; the last two species originate from the volumetric pyrolysis reaction. The conservation equations are derived from the Boltzmann equations through ensemble averaging. Stresses in the gaseous phase are the sum of the Newtonian and Reynolds (turbulent) contributions. The particulate phase stresses are the sum of collisional and Reynolds contributions. Heat transfer between phases, and heat transfer between classes in the particulate phase is modeled, the last resulting from collisions between sand and biomass. Closure of the equations must be performed by modeling the Reynolds stresses for both phases. The results of a simplified version (first step) of the model are presented.

1. INTRODUCTION

Fluidized beds are cylindrical, vertical containers that are extensively used in industry for chemical conversion of substances from solid particles. In a 'bubbling' fluidized bed, a solid particulate phase undergoes a chemical reaction whose products are carried away by a flow introduced through openings in the bottom section of the reactor. The 'bubbles' are regions of the mixture mostly devoid of particles; these regions occur as a result of the interaction of the turbulent gaseous flow with the particles. This investigation is devoted to the study of such a fluidized bed whose particulate phase consists of initially room-temperature biomass particles mixed with hot sand; the sand is a high heat capacity medium whose role is to heat the biomass which pyrolyses upon reaching a threshold temperature [1].

Despite the many models of fluidized beds, Kumaran [2] recently acknowledges that there is still no consensus on an appropriate continuum description. According to Enwald et al. [3], few of the existing models have been satisfactorily validated and even when validated, they contain assumptions that may invalidate their extension to general fluidized beds. For example, the Boussinesq approximation used to model gas phase turbulence

is invalid [4] for modeling flows such as those in fluidized beds which exhibit regions of recirculating flow. The *status quo* is such that the majority of the models are descriptive rather than predictive. Our goal is to derive a model that has predictive capabilities.

Lahey and Drew [5] have proposed a consistent formalism for the description of granular flows with no chemical reaction based upon the concepts of ensemble averaging and phase averaging. Additionally, Drew [6] included particle-velocity fluctuations effects in the previously derived model to account for their influence on the interfacial force and stresses. Prosperetti et al. [7] and Zhang and Prosperetti [8] have derived a model based on a probabilistic approach combined with phase averaging; these models are primarily focussed on the continuity and momentum equations, and [7] considers the simpler case of an incompressible carrier flow. An interesting class of models is that of Simonin and co-workers [9–13] for isothermal mixtures without phase change, and [14] for a more general case but without chemical reactions. The same model has been adopted by Enwald et al. [3] and by Peirano [15]. These models are derived by taking moments of the Boltzmann equation, and the most advanced [12–15] include an extension of the Jenkins and Richman [18] model for internal stress calculation to cases where there is a carrier flow interacting with the particles. Louge et al. [19,20] have also included the influence of gas-particle interactions in the collisional stresses but they do not model collisional heat transfer.

The model derived below follows the spirit of Simonin [14] and Peirano [15] and additionally includes: (1) a general model of volumetric particle-reaction, (2) species equations for both gas and solid, (3) a formulation for the usually neglected stress tensor resulting from different-class particle collisions, (4) an explicit model of the heat transfer resulting from different-class particle collisions, and (5) a turbulence model for the carrier phase (interacting with the particles) that is not based upon the Boussinesq approximation. Although Jenkins and Mancini [21,22] have modeled binary mixtures of particles, there is no current model including all of the above phenomena.

2. MODEL DESCRIPTION

The model derivation parallels that of Chapman and Cowling [23] for the conservation equations in a dense gas with the differences that here a gas interacts with the particles, that collisions may be inelastic, that particles mass may change, etc. The particulate phase is divided into M classes of particles (here $M = 2$: biomass, b , and sand, s) and a distribution function is defined for each class $f_j(\mathbf{u}_j, m_j, T_j, Y_{i,j}; \mathbf{x}, t)$ with $j \in [1, M]$ and $i \in [1, N_j]$ (number of species of class j) where $\mathbf{u}_j, m_j, T_j, Y_{i,j}$ are respectively the velocity, mass, temperature and species i mass fraction in class j , and \mathbf{x} is the location of class j particles at time t . By definition, $n_j(\mathbf{x}, t) = \int f_j(\mathbf{u}_j, m_j, T_j, Y_{i,j}; \mathbf{x}, t) d\boldsymbol{\xi}_j$ ($d\boldsymbol{\xi}_j \equiv d\mathbf{u}_j dm_j dT_j dY_{i,j}$) is the number of particles of class j per unit volume at any location \mathbf{x} in the physical domain. Both ensemble averages and Favre ensemble averages are defined for a property ψ_j

$$\overline{\psi_j} \equiv (n_j)^{-1} \int \psi_j f_j d\boldsymbol{\xi}_j, \quad \overline{\psi_j'} \equiv \overline{\psi_j - \overline{\psi_j}}; \quad \overline{\overline{\psi_j}} \equiv \overline{m_j \overline{\psi_j}} / \overline{m_j} = n_j \overline{m_j \overline{\psi_j}} / (\alpha_j \overline{\rho_j}), \quad \psi_j'' \equiv \psi_j - \overline{\overline{\psi_j}} \quad (1)$$

where $\alpha_j \equiv n_j \overline{m_j} / \overline{\rho_j}$. The particulate phase conservation equations are derived by multiplying the Boltzmann equation by $m_j \psi_j$; performing ensemble averages for $\psi_j = m_j^{-1}$

(number density), 1 (continuity), \mathbf{u}_j (momentum), $C_{pj}T_j$ (enthalpy), and $Y_{i,j}$ (species); and replacing in the resulting equations the source terms of the form $d\psi_j/dt$ with the expressions found from the Lagrangian, single particle dynamics equations. The gas phase conservation equations are the classical Navier-Stokes (NS) equations.

2.1. Particle conservation equations

Particle number density:

$$\partial(n_j)/\partial t + \nabla \cdot (n_j \overline{\mathbf{u}}_j) = 0 \quad (2)$$

assuming that there is no particle coalescence or break-up (no source/sink) during collisions.

Continuity:

$$\partial(\alpha_k \overline{\rho}_k)/\partial t + \nabla \cdot (\alpha_k \overline{\rho}_k \overline{\mathbf{u}}_k) = \alpha_k \overline{\rho}_k \overline{[(1/m_k) dm_k/dt]} \quad (3)$$

assuming that there is no mass exchange during particle collisions.

Momentum:

$$\begin{aligned} \partial(\alpha_k \overline{\rho}_k \overline{\mathbf{u}}_k)/\partial t + \nabla \cdot (\alpha_k \overline{\rho}_k \overline{\mathbf{u}}_k \overline{\mathbf{u}}_k) = & -\nabla \cdot (\alpha_k \overline{\rho}_k \overline{\mathbf{u}}_k'' \overline{\mathbf{u}}_k'') + \alpha_k \overline{\rho}_k \overline{d\mathbf{u}}_k/dt \\ & + \alpha_k \overline{\rho}_k \overline{[(\mathbf{u}_k/m_k) dm_k/dt]} + \mathbf{C}_k(m_k \mathbf{u}_k) \end{aligned} \quad (4)$$

where $\overline{\mathbf{T}}_k^{\text{Re}} \equiv -\overline{\mathbf{u}}_k'' \overline{\mathbf{u}}_k''$ are the Reynolds stresses which must be modeled, and $\mathbf{C}_k(m_k \mathbf{u}_k)$ are the collisional (viscous) stresses augmented by contributions (sources) from collisions between different-class (i.e. sand-biomass) particles.

Enthalpy:

$$\begin{aligned} \partial(\alpha_k \overline{\rho}_k \overline{C_{pk} T_k})/\partial t + \nabla \cdot (\alpha_k \overline{\rho}_k \overline{\mathbf{u}}_k \overline{C_{pk} T_k}) = & -\nabla \cdot (\alpha_k \overline{\rho}_k \overline{\mathbf{u}}_k'' \overline{C_{pk} T_k'')}) + \alpha_k \overline{\rho}_k \overline{C_{pk} dT_k/dt} \\ & + \mathbf{C}_k(m_k C_{pk} T_k) + \alpha_k \overline{\rho}_k \overline{[(C_{pk} T_k/m_k) dm_k/dt]} \end{aligned} \quad (5)$$

where $\mathbf{C}_k(m_k C_{pk} T_k)$ is the collisions generated heat flux/source; we assume a null reference T in the definition of the enthalpy.

Species (for $k = b$ only):

$$\begin{aligned} \partial(\alpha_k \overline{\rho}_k \overline{Y_{i,k}})/\partial t + \nabla \cdot (\alpha_k \overline{\rho}_k \overline{\mathbf{u}}_k \overline{Y_{i,k}}) = & -\nabla \cdot (\alpha_k \overline{\rho}_k \overline{\mathbf{u}}_k'' \overline{Y_{i,k}'}) + \alpha_k \overline{\rho}_k \overline{dY_{i,k}/dt} \\ & + \alpha_k \overline{\rho}_k \overline{[(Y_{i,k}/m_k) dm_k/dt]} \end{aligned} \quad (6)$$

2.2. Gas conservation equations

The procedure for obtaining the gas conservation equations is to multiply the single phase NS equations by a phase indicator, χ , that is defined as being unity in the gas and null otherwise, and therefore moves with the velocity of receding solid surfaces, \mathbf{u}_l , due to pyrolysis. These equations are then ensemble averaged (symbol $\langle \rangle$) over all realizations of NS variables, and new averaged variables and their fluctuations are defined; both regular ($\overline{\psi}$) and Favre ($\widetilde{\psi}$) averages are defined. The average of the phase indicator is denoted by α_g .

Continuity:

$$\partial(\alpha_g \bar{\rho}_g)/\partial t + \nabla \cdot (\alpha_g \bar{\rho}_g \tilde{\mathbf{u}}_g) = \langle \rho_g(\mathbf{u}_g - \mathbf{u}_l) \cdot \nabla \chi_g \rangle \quad (7)$$

Momentum:

$$\begin{aligned} \partial(\alpha_g \bar{\rho}_g \tilde{\mathbf{u}}_g)/\partial t + \nabla \cdot (\alpha_g \bar{\rho}_g \tilde{\mathbf{u}}_g \tilde{\mathbf{u}}_g) &= \nabla \cdot \left[\alpha_g \left(\overline{\overline{T}_g} + \overline{\overline{T}_g^{\text{Re}}} \right) \right] + \alpha_g \bar{\rho}_g \tilde{\mathbf{F}}_g + \\ &\quad - \overline{\overline{T}_g} \cdot \nabla \chi_g + \langle \rho_g(\mathbf{u}_g - \mathbf{u}_l) \mathbf{u}_g \cdot \nabla \chi_g \rangle \end{aligned} \quad (8)$$

Species:

$$\begin{aligned} \partial(\alpha_g \tilde{Y}_{i,g} \bar{\rho}_g)/\partial t + \nabla \cdot (\alpha_g \bar{\rho}_g \tilde{Y}_{i,g} \tilde{\mathbf{u}}_g) &= -\nabla \cdot (\alpha_g \overline{\mathbf{J}_{i,g}}) + \overline{S_{i,g} \alpha_g} - \nabla \cdot (\alpha_g \bar{\rho}_g \widetilde{Y_{i,g}'' \mathbf{u}_g''}) \\ &\quad - \overline{\mathbf{J}_{i,g}} \cdot \nabla \chi_g + \langle \rho_g Y_{i,g}(\mathbf{u}_g - \mathbf{u}_l) \cdot \nabla \chi_g \rangle \end{aligned} \quad (9)$$

Energy:

$$\begin{aligned} \partial[\bar{\rho}_g \alpha_g \tilde{e}_{Tg}]/\partial t + \nabla \cdot [\bar{\rho}_g \alpha_g \tilde{e}_{Tg} \tilde{\mathbf{u}}_g] &= \sum_i \overline{S_{i,g} \Delta h_i \alpha_g} + \alpha_g \tilde{\mathbf{F}}_g \cdot \tilde{\mathbf{u}}_g - \nabla \cdot [\bar{\rho}_g \alpha_g \widetilde{e_{Tg}'' \mathbf{u}_g''}] \\ &\quad + \alpha_g \overline{\mathbf{F}_g'' \cdot \mathbf{u}_g''} - \nabla \cdot [\alpha_g (\overline{\mathbf{q}_g} - \overline{\overline{T}_g} \cdot \tilde{\mathbf{u}}_g - \overline{\overline{T}_g} \cdot \mathbf{u}_g'')] \\ &\quad + \langle \left[\rho_g(\mathbf{u}_g - \mathbf{u}_l) e_g + (\mathbf{q}_g - \overline{\overline{T}_g} \cdot \mathbf{u}_g) \right] \cdot \nabla \chi_g \rangle \end{aligned} \quad (10)$$

where the right hand side of eq. 7 is the averaged mass flux at surfaces l ; the last term in eq. 8 is the interfacial averaged momentum due to the mass flux at surfaces l ; the third term in the right hand side of eq. 9 is the double correlation between species and velocity fluctuations, the fourth term is the averaged mass diffusion flux of species i at surfaces l , and the fifth term is the interfacial averaged flux of species i at surface l ; the term involving $\overline{\overline{T}_g} \cdot \mathbf{u}_g''$ in eq. 10 contains the pressure-velocity correlation and the Reynolds stress shear work, and the terms in the last line represent respectively the energy evolved at surfaces due to mass evolution, the contact heat transfer evolution and work due to drag. All of the surface contribution terms must be modeled.

2.3. Modeling of the interphase transfer and reaction source/sink terms

2.3.1. Interphase transfer

The biomass reaction scheme adopted here is that of Miller and Bellan [1] in which there are initially (virgin species) three components (cellulose, hemicellulose and lignin) following identical reaction paths, but with different (first order) rates. Each component β depolymerizes to an 'active' species (reaction rate K_1^β) which further reacts to yield tar (K_2^β), and char and gas (K_3^β) in the proportion $X^\beta/(1 - X^\beta)$. Additionally, tar reacts to form gas (K_4^β). The Lagrangian equations contain explicitly the interphase transfer terms as follows:

Species: $d(\rho_b Y_{act,b}^\beta)/dt = S_{act \rightarrow g}^\beta + S_{act \rightarrow vir}^\beta$ where $S_{act \rightarrow g}^\beta$ is the interphase production while $S_{act \rightarrow vir}^\beta$ is the in-phase reaction (see below). $S_{act \rightarrow g}^\beta = -\rho_{act}^\beta K_2^\beta - (1 - X^\beta)\rho_{act}^\beta K_3^\beta$ for each β , where $K_i = A_i^\beta \exp[-E_i^\beta/R_u T_b]$ with the Arrhenius parameters A_i^β and E_i^β determined by Miller and Bellan [1]. The corresponding terms in the mass fraction equations for the gas phase are $S_{tar \rightarrow act}^\beta = \rho_{act}^\beta K_2^\beta$ and $S_{gas \rightarrow act}^\beta = (1 - X^\beta)\rho_{act}^\beta K_3^\beta$.

Mass: $d\rho_b/dt = \sum_\beta S_{act \rightarrow g}^\beta$

Momentum: $du_k/dt = \mathbf{F}_k/m_k - (\nabla \hat{p}_g)/\rho_k - C_D 0.5\pi R_k^2 (\mathbf{u}_k - \hat{\mathbf{u}}_g) |\mathbf{u}_k - \hat{\mathbf{u}}_g|$ where \mathbf{F}_k is a body force, \hat{p}_g and $\hat{\mathbf{u}}_g$ correspond to the turbulent, but undisturbed-by-the-particle values, C_D is the drag coefficient that incorporates the blowing effect from the particle (Bellan and Harstad, 1987 [16]) and R_k is the particle radius. In this model $\mathbf{F}_s = \mathbf{g}$ under the assumption of particle symmetric mass transfer.

Enthalpy: $m_k C_{pk} dT_k/dt = \sum_r S_{rk} \Delta h_{rk} + Q_{kg} + \sum_{s \rightarrow g} \dot{m}_{ki} h_i$, where r denotes reaction, Δh is a heat of reaction (values provided in [1]), h_i is the heat convected away with the mass of each species i leaving the particle, $S_{rs} = 0$, and $Q_{kg} = \lambda_g 2\pi R_k Nu(\text{Re}_g, \text{Pr}_g)(\hat{T}_g - T_k)$ is the conductive/convective heat transfer where $\text{Re}_g = 2R_k |\hat{\mathbf{u}}_g - \mathbf{u}_k|/\nu_g$ and $\text{Pr}_g = \rho_g \nu_g C_{pg}/\lambda_g$; λ_g is the gas thermal conductivity, and ν_g is the gas kinematic viscosity.

2.3.2. In-phase reaction source/sink terms

Reactions K_1, K_2 and K_3 convert species within the same phase [1]; this occurs respectively for the 'active' species originating from the virgin species, $S_{act \rightarrow vir}^\beta = -\rho_{vir}^\beta K_1 = -S_{act \rightarrow vir}^\beta$, for the char resulting from the 'active' species, $S_{char}^\beta = X^\beta \rho_{act}^\beta K_2$, and for the gas, $S_{gas}^\beta = \rho_{tar}^\beta K_4$.

2.4. Modeling of the collisional terms

2.4.1. Collisional particulate stresses

Jenkins and Savage [17] were the initiators of a comprehensive model of particle collisions originated stresses in the simple case of single class particles in absence of an interstitial gas; this model was further elaborated by Jenkins and Richman [18]. Louge et al. [19,20] have extended this model to steady gas-particle interactions, while Simonin [14] and Peirano [15] have included the effect of gas phase turbulence and influence of turbulence on particle drag. Furthermore, Jenkins and Mancini [21,22] have modeled collisions in binary particles mixtures, but without gas interaction. The present model is inspired by the preceding work and incorporates turbulence, gas-solid interactions and collisions between different-class particles. Here, the theory of Jenkins and Mancini [21] is used for a mixture of binary (sand-biomass), nearly elastic spheres, but the momentum equation includes now the drag term, just as the formalism of Simonin [14] and Peirano

[15], accounts for the gas-particle interactions.

2.4.2. Collisional heat transfer

To our knowledge, collisional heat transfer between particles has never been modeled in the context of fluidized beds. Collisional heat transfer may in principle occur between like (sand-sand or biomass-biomass) and unlike (sand-biomass) particles. In either situation, a model of collisional heat transfer can be obtained only if one knows three quantities: (1) the energy transferred during a collision, ΔE_{coll} , (2) the contact time τ_c , and (3) the collisional area, A_{coll} . For two particles A and B of different temperatures T_A and T_B , one may estimate $\Delta E_{coll} = 2A_{coll}(\tau_c^{0.5}/\pi)[\omega_A\omega_B(T_A - T_B)/(\omega_A + \omega_B)]$ where $\omega_I = [\lambda_I/(\rho_I C_{p,I})]^{0.5}$. The collisional time is here modeled under the assumption that heat transfer arises from the sliding contact of two particles yielding $\tau_c = \kappa d / |\mathbf{u}_A - \mathbf{u}_B|$, where d is the diameter of the large particle and κ is an empirical constant to be determined by comparing with appropriate experiments. The collisional area is assumed to be determined by the size of the smaller particle. Using these quantities, the theory of Jenkins and Mancini [21] is used to calculate the source and flux terms of ΔE_{coll} .

2.5. Modeling of the Reynolds stresses

2.5.1. Carrier gas

Unlike most models of gas turbulence in fluidized beds, our model does not contain the Boussinesq approximation which implies that the stress and strain rate tensors are aligned. This assumption is not consistent with recirculating flow, which is one aspect of fluidized beds [4]. Even with the Boussinesq approximation, numerical results may show recirculating zones because of the boundary conditions, but the results are inconsistent with the assumptions and therefore their validity is suspicious. Here, we model the carrier turbulence using the full differential Reynolds Stress Model (DRSM) equations for two phase flows. The DRSM equations are obtained from the conservation equations by calculating the average of $[\chi u_i'' NS(u_j) + \chi u_j'' NS(u_i)]$ where $NS(u_i)$ represents the i th component of the momentum equations. The general result is:

$$\partial < \chi \rho u_i'' u_j'' > / \partial t + \partial [\tilde{u}_k < \chi \rho u_i'' u_j'' >] / \partial x_k = P_{ij} + T_{ij} + \Pi_{ij} + \varepsilon_{ij} + W_{ij} + M_{ij} \quad (11)$$

where all right hand side (RHS) terms of the equation, except the first, need modeling. The RHS terms represent respectively production due to gradients in mean velocity, transport due to both molecular and turbulent processes, the pressure-strain correlation, energy dissipation, interfacial work due to exchanges across the interface, and effect of mass transfer across the interface.

2.5.2. Particles

The terminology used in [18] for these stresses is the ‘between collisions’ stresses, as they are due to correlations between particle velocity fluctuations. Following Simonin [14] and Peirano [15], the Boussinesq approximation is used to model the turbulent stresses: the practical implication is that the granular temperature (proportional to the particle turbulent kinetic energy) is sufficient to describe the turbulent features of the particulate flow. The dissipation rate entering the kinetic energy equation is here calculated with the expressions obtained from the collisional model of Jenkins and Mancini [22]. This

approximation will be checked *a posteriori* by comparing the predictions of the numerical simulation with data.

3. SUMMARY

A model of biomass pyrolysis in a fluidized bed reactor has been (due to space constraints) briefly described. The model is extremely complex due to the coupled nature of the dynamic and thermodynamic evolution of the phases, and also to the turbulent features of the carrier and particulate flow. Since many of the aspects of the model are novel, it is prudent to validate them sequentially. For this reason, the first set of results to be presented is that from simulations of the sand-biomass dynamics in absence of heat transfer, turbulence or reaction. The results thus obtained allow the validation of both the dynamic interaction between phases and the calculation of the particulate stress tensor.

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